

AMENDMENTS TO THE SPECIFICATION

Please replace paragraph [0073] with the following amended paragraph:

[0073] The temperature in furnace 303 may be reduced at a predetermined rate (for example, 1°C per minute) by furnace controller 307 wherein the furnace temperature is measured by temperature sensor 305 and a temperature signal is transmitted to controller 307. The temperature also is measured by temperature sensor 355, via line 357, and compared with the stored isocompositional P_{O_2} - temperature equilibrium relationship (e.g., Fig. 1), the appropriate P_{O_2} is determined, and the controller sends the proper set points via signal lines 343 and 345 to flow controllers 313 and 319, respectively. The relative flows of air and fuel via lines 315 and 321 to burner 323 and the total pressure in furnace 303 are controlled so that the P_{O_2} is substantially in equilibrium with the stoichiometric composition of the mixed conducting metal oxide material in articles 301 at the measured temperature. The total pressure in the furnace may be maintained at a constant level by controller 339 via pressure control valve 327 and/or backpressure control valve 333. Control by properly decreasing P_{O_2} is continued in this manner as the temperature is reduced to about 100°C, below which control of P_{O_2} may not be necessary.

Please replace paragraph [0059] with the following amended paragraph:

[0059] The oxygen-containing gas is provided in line 211 at a selected flow rate controlled by flow controller 213 and flows through line 215. Diluent gas is provided in line 217 at a selected flow rate controlled by flow controller 219 and flows through line 221. The oxygen-containing gas and diluent gases may be combined to provide ~~[[a]]~~ an oxygen-containing gas that flows via line 223 into furnace 203. Vent gas flows from the furnace via line 225, passes through optional gas analyzer 227 and flow control or backpressure regulating valve 229, and may be vented to the atmosphere via line 231 or sent to a vent gas recovery system (not shown) if desired. The gas pressure in furnace 203 may be controlled by pressure control valve 233 and/or regulating valve 229.

Please replace paragraph [0074] with the following amended paragraph:

[0074] As an alternative to or in addition to combusting air and fuel over the entire cooling temperature range, when the gas pressure in furnace 303 is above atmospheric pressure, the total pressure in the furnace may be reduced by controller 339 via pressure control valve 327 and/or backpressure control valve ~~[[335]]~~ 335 as the temperature drops during cooling, thereby controlling P_{O_2} to maintain an isocompositional state in articles 301. This control of P_{O_2} via total pressure control will be effective down to a certain temperature, below which the gas composition must be controlled as described above for the remainder of the cooling.

Please replace paragraph [0079] with the following amended paragraph:

[0079] Membrane module 401 typically operates in the temperature range of 650 to 1000°C and may be heated and maintained at temperature by heating oxygen-containing gas in line 409 in an upstream heating system. In the illustration of Fig. 4, an oxygen-containing gas (for example, air) via line 416 and a fuel (for example, natural gas) via line 417 are combusted in burner 419 to provide the hot oxygen-containing gas in line 409 that is introduced into membrane module 401. Optionally, a diluent (for example, nitrogen or a portion of the O_2 -depleted non-permeate from line 411) may be introduced via line 421 and/or line 415. Alternatively, an oxygen-rich gas (i.e., a gas containing a higher oxygen concentration than the O_2 -containing gas in line 409) may be introduced via line 421 and/or line 415. Optionally, indirectly-fired burner or process furnace 423, which is fired by a fuel (for example, natural gas) via line 425 and ~~[[a]]~~ an oxygen-containing gas (for example, air) via line 427, may be used to further heat the hot oxygen-containing gas from burner 419 without changing the oxygen partial pressure of this gas.

Please replace paragraph [0092] with the following amended paragraph:

[0092] During startup, P_{O_2} must be controlled in each of the modules 501, 505, 509, and 513 and this may not be possible if P_{O_2} is controlled only in the oxygen-containing gas feed in line 409. During heating, a temperature profile may exist in the axial direction (i.e., the flow direction from the inlet of module 501 to the outlet of module ~~[[313]]~~ 513). Depending on the membrane composition, oxygen permeation may start to be significant at a temperature as low as 400°C. If the oxygen partial pressure is not controlled on both sides of the membranes 501c, 505c, 509c, and 513c, oxygen may permeate from one side of the membrane to the other. If this occurs, the P_{O_2} will be different in the oxygen-containing feed gas side of each module.

Please replace paragraph [0094] with the following amended paragraph:

[0094] In order to control the P_{O_2} properly on the permeate sides of the membrane modules ~~[[501 through 513]]~~ 501, 505, 509 and 513 to minimize deviation from the isocompositional curve of Fig. 1, gas of controlled composition may be introduced into each module via lines 517, 519, 521 and 523. The composition of the gas would be chosen such that the gas phase composition is substantially in chemical equilibrium with the oxygen stoichiometry of the membrane. The flow rate of the gas would be chosen to provide sufficient time for the gas phase composition to equilibrate relative to the changing gas-phase temperature.

Please replace paragraph [0098] with the following amended paragraph:

[0098] In order to control P_{O_2} properly on the interior of the membrane modules ~~[[501 through 513]]~~ 501, 505, 509 and 513 to minimize deviation from the isocompositional curve of Fig. 1, gas of controlled composition may be introduced into each module via lines 517, 519, 521 and 523. The composition of the gas would be chosen such that the gas phase composition is substantially in chemical equilibrium with the oxygen stoichiometry of the membrane. The flow rate of the gas would be chosen to provide sufficient time for the gas phase composition to equilibrate relative to the changing gas-phase temperature. The composition of the gases in the oxygen-containing feed side and the permeate side of each membrane may be independently controlled to maintain an oxygen stoichiometry gradient across the membrane as the module is cooled or heated. This gradient may be

the same oxygen stoichiometry gradient that exists across the membrane during steady state operation at the operating temperature.

Please replace paragraph [0099] with the following amended paragraph:

[0099] An alternative embodiment of this technique would control the total pressure inside each module as the temperature is changed. For example, at steady state operating temperature, the total pressure inside the modules is set to the desired value by withdrawing gas through line 525. As the system is shut down and the temperature decreased, the total pressure would be decreased via ~~[[lines]]~~ line 525 to decrease the oxygen partial pressure inside each module.

Please replace paragraph [0101] with the following amended paragraph:

[0101] Preheated reactant gas (for example, a methane-containing gas such as natural gas or a prereformed natural gas/steam/synthesis gas mixture) is introduced via line 617 and flows in series through the reactant sides of modules 601, 603, 605, and 607 while reacting therein with oxygen that has permeated through membranes 601b, 603b, 605b, and 607b, respectively. Reaction product gas (for example, synthesis gas comprising hydrogen and carbon monoxide) is withdrawn from the system via line 619. During steady-state operating conditions, the temperature and/or the stoichiometric composition of the membranes may differ in the axial or flow direction from module 601 to module 607. When the system is shut down, cooling will begin with these profiles and P_{O_2} must be controlled separately in each module during at least part of the cooling period.

Please replace paragraph [0102] with the following amended paragraph:

[0102] The terms "series" and "series operation" as used herein means that a process stream leaving any module flows into the next module in the series of modules. In this embodiment, the reactant gas leaving one module provides the ~~provides the~~ reactant gas flowing to the next module.

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Please replace paragraph [0104] with the following amended paragraph:

[0104] Alternatively, during system startup, the oxygen partial pressure in oxygen-containing feed gas sides 601c, 603c, 605c, and 607c may be controlled by blending an oxygen-containing gas such as air from line 409 with a diluent gas via lines 621, 623, 625, and 627, respectively. The total pressure of the oxygen-containing gas in oxygen-containing feed gas sides 601c, 603c, 605c, and ~~[[670c]]~~ 607c may be controlled by known means as necessary during heating, steady state operation, and cooling of the module system.